The Quasi-1D S=1/2 Antiferromagnet Cs_2CuCl_4 in a Magnetic Field

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Abstract

Magnetic excitations of the quasi-1D $S = 1/2$ Heisenberg antiferromagnet (HAF) Cs_2CuCl_4 have been measured as a function of magnetic field using neutron scattering. For $T < 0.62$ K and B=0 T the weak inter-chain coupling produces 3D incommensurate ordering. Fields greater than B_C =1.66 T, but less than the field ($\simeq$ 8 T) required to fully align the spins, are observed to decouple the chains, and the system enters a disordered intermediate-field phase (IFP). The IFP excitations are in agreement with the predictions of Müller *et al.* for the 1D $S = 1/2$ HAF, and Talstra and Haldane for the related $1/r^2$ chain (the Haldane-Shastry model). This behaviour is inconsistent with linear spin-wave theory.

75.10.Jm, 75.10.Hk, 75.30.Ds

In recent years there has been a general realization that widely used semi-classical and mean-field theories of magnetism, such as linear spin-wave theory, are often inadequate in describing the properties of low-dimensional quantum magnets [1]. Research on 1D systems has benefited from a strong interaction between theory and experiment, with novel concepts such as fractional excitations and topological energy gaps being introduced and developed. New magnetic materials continue to be found and the body of experimental and theoretical work is increasing. In this Letter we describe a further contribution to this progress by presenting experiments which show the novel features which occur when a magnetic field is applied to a quasi-1D antiferromagnet.

The canonical 1D quantum mechanical system is the $S = 1/2$ Heisenberg antiferromagnetic chain (HAFC). The low-temperature ground state is a non-magnetic spin singlet [2], and the excitations are spin-$\frac{1}{2}$ particles known as spinons, as shown by the solution of the $1/r^2$ Haldane-Shastry model (HSM) [3]. In a neutron-scattering process the spin of the system changes by 1 or 0, and so the neutron-scattering measures a spinon pair continuum. Observations of this continuum for quasi-1D HAFC's have been made and confirm the spinon picture [4]. The application of a magnetic field aligns a number of the spins along the field by creating an approximately regular array of condensed spinons. The triplet excitation continuum in zero field is then split into separate continua [5,6] as illustrated in Fig. 1. The positions of the continua alter as the applied field increases until at high fields $B \geq B_0$ the spins are aligned parallel along the field and the excitations have a well-defined magnon dispersion [5].

The $S = 1/2$ HAFC has also been treated by mapping the spin chain into a system of interacting fermions [7]. In zero field the fermion band is half full and the excitation energies can be obtained using the Hartree-Fock approximation. The transverse excitations are well-defined and have dispersion relations similar to the lower boundaries (heavy lines) of Fig. 1(a). The application of a field changes the chemical potential and the Fermi wavevector, such that the transverse excitations have incommensurate dispersion relations similar to the heavy lines in Fig. 1(b).

Although there are a number of excellent realizations of the $S = 1/2$ HAFC available, Cs_2CuCl_4 is one of the few quasi-one-dimensional systems with a *sufficiently low exchange interaction* that studies of an applied field *throughout* the intermediate-field phase (IFP) can be made. The IFP is the region of the phase diagram which occurs at low temperatures ($T \ll J/k_B$), for fields below the ferromagnetic transition field B_0, and when the chains behave independently. For a quasi- as opposed to pure-1D antiferromagnet this is above a critical field B_C, the field necessary to decouple the chains, and the IFP region occurs for $B_C < B < B_0$.

Measurement of magnetic excitations in the IFP is technically challenging as it requires intense beams of cold neutrons, high magnetic fields, low temperatures and large single crystals of a quasi-1D material with a low exchange. Because of this, previous experiments on quasi-1D $S = 1/2$ HAFC's have been exclusively in the province of applied fields which are low with respect to the exchange, and the difficulty of these experiments has precluded a comparison with theory [8]. In this Letter we report neutron scattering measurements which, for the first time, span a large region of the IFP. Although Cs_2CuCl_4 has a large inter-chain coupling ($\sim 17\%$ of the intra-chain value) we argue that the field decouples the chains and good agreement is found with the theories of Müller *et al.* [5], and Talstra and Haldane [6].

Cs_2CuCl_4 has an orthorhombic crystal structure [9]. The $S = 1/2$ Cu^{2+} spins are coupled into chains running parallel to the b-axis, with four such chains passing through each unit cell. The magnetic susceptibility [10] is consistent with a quasi-1D $S = 1/2$ HAFC with an interaction $J = 0.34 \pm 0.02$ meV, where the exchange Hamiltonian for the 1D chains is $\mathcal{H}_{1D} = J \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1}$. The chains are coupled in the c-direction by a small exchange $J' = 0.175J$ as depicted in Fig. 2(a) (all other inter-chain exchanges are negligible) [11]. Below T_N= 0.62 K the spins order into a cycloid along the chain direction with an incommensurate wavevector $\mathbf{q}$=(0,0.472,0); the incommensurate ordering is due to the frustration caused by the staggering of chains with respect to their neighbours, see Fig. 2(a). A small anisotropy confines the spins to rotate within a plane containing the b-direction and making a small

angle with the (b, c) plane.

The single crystal of Cs_2CuCl_4 [11] was mounted with the (a, b) scattering plane horizontal and a magnetic field was applied vertically. The sample was cooled using either a dilution refrigerator or a ^{3}He cryostat which provided base temperatures of 0.06 or 0.32 K, respectively. The inelastic neutron scattering experiments were performed on the three-axis crystal spectrometer IN14 at Institut Laue-Langevin, Grenoble, France, while the elastic scattering measurements were made with the three-axis crystal spectrometer TAS7 at Risø National Laboratory, Denmark. Full details of the experiments will be given elsewhere [12].

Strong magnetic reflections were observed at B=0 T and T=0.32 K in the (a, b) plane at positions corresponding to the cycloidal ordering wavevector $\mathbf{q}$=(0,0.472,0). When a field was applied along the crystallographic c-axis the intensity of the magnetic Bragg peaks decreased with increasing B and finally disappeared at the critical field B_C=1.66 T (T=0.32 K); for all fields and temperatures the incommensurate ordering wavevector remained constant. The observed transition field is much smaller than the field B_0 necessary for full ferromagnetic alignment which was found to be $B_0 \simeq 8$ T from susceptibility measurements [12]. Fig. 2(b) shows a summary of the magnetic phase diagram for Cs_2CuCl_4. No antiferromagnetic or incommensurate Bragg peaks were detected in the intermediate-field phase (IFP), suggesting a decoupling of the chains.

Fig. 2(c) shows the dispersion curves measured at T=0.1 K in zero applied field. The asymmetry in the dispersion relation arises from the inter-chain coupling and is well accounted for by a calculation of the spin waves with a Heisenberg exchange J for the interactions along the chain and J' for the coupling between chains [13]. The solid line in Fig. 2(c) represents a fit of the dispersion relation with $J = 0.64 \pm 0.02$ meV and $J'/J = 0.17 \pm 0.02$. The latter ratio of exchange interactions is in good agreement with the value $J'/J = 0.175$ which gives a stable incommensurate structure [11]. The magnitude of J is larger than the value 0.34 meV deduced from the susceptibility measurements [10] by using a theory which includes quantum fluctuations. The quantum correction to the classical spin-wave result for

an ideal $S = 1/2$ HAF chain is a multiplication of the energy scale by a factor of $\frac{\pi}{2}$ [14], and a value of $\tilde{J} = \frac{2}{\pi}$ 0.64=0.41 meV is similar to the value deduced from susceptibility measurements.

The magnetic excitations were studied as a function of field by measuring the inelastic scattering at low temperatures (T=0.06 K). Fig. 3 summarizes the results of constant-Q scans for Q=(0,0.75,0). The zero-field scan shows a well-defined, almost resolution-limited peak centered at E=0.58 $\pm$ 0.01 meV. The solid line is a fit to a Gaussian peak and the tail at low energies comes from the incoherent scattering. The intensity of the magnetic excitation decreases on increasing the field and the lineshape changes above the IFP transition at $B_{\rm C}$=1.66 T. A good but not unique fit to the lineshape is obtained by two independent peaks, as shown by the solid line in the figure. The low-energy peak decreases in energy on increasing the field and has an almost constant width and intensity for $B_C < B < 3$ T. In the same field range, the high-energy peak is at E=0.66 $\pm$ 0.02 meV, slightly increased in energy compared to the peak position in the 3D cycloidal phase. It becomes broader on increasing the field, but its total integrated intensity is almost constant.

The inelastic scattering in fields up to 6 T was also measured at Q=(-0.25,2,0), which is equivalent to the zone center for a quasi-1D magnetic system with chains along b^*. The results show an inelastic peak for $B > 3$ T which increases in energy and becomes more intense on increasing the field. This inelastic peak measures the Zeeman energy gap $E_Z = g_c \mu_B B$, where g_c is the g-value along the c-axis. A fit to the peak positions gives $g_c = 2.36 \pm 0.02$, which is in good agreement with the value $g_c = 2.30 \pm 0.01$ determined by previous room temperature EPR measurements [15].

The results of scanning Q across the zone with E=0.35 meV are presented in Fig. 4. The zero-field scan has inelastic peaks occurring at positions corresponding to the intersection of the scan direction with the dispersion relation, Fig. 2(c). The magnetic inelastic scattering decreases in intensity and becomes broader on increasing field. The double-peak structure at the zone boundary k=1.5 transforms into a broad feature and the peak on the low-Q side is displaced towards the zone boundary and cannot be distinguished from the other peak

for $B > 4$ T.

Mean-field ordering and linear spin-wave theory predict that in the case of a field applied in the plane of spin rotation the IFP is either a spin-flop (SF) or a cone structure. Both of these have long-range antiferromagnetic-like order which was not observed. If the anisotropy confining the spins in the (b, c) plane is relatively large, the structure is a SF phase with antiferromagnetic ordering along b, and the transition field cycloidal $\rightarrow$ SF phase is calculated to be B_C=1.425 T [16]. The excitation energy at Q=(0,0.75,0) for isolated 1D chains is field-independent and equal to 0.64 meV [5]. This is clearly inconsistent with the observed splitting of the inelastic scattering as a function of field, see Fig. 3. Inter-chain coupling causes some broadening of the scattering, but cannot account for the results.

In the same mean-field approach, for smaller anisotropies the magnetic field produces a cone phase in which the spins cant towards the field direction to gain energy through the Zeeman interaction and the transverse spin components rotate in the (a, b) plane with the same wavevector $\mathbf{q}$ as the zero-field structure in order to minimize the exchange interactions [16]. The spin-waves in the cone phase have been calculated [17] and the results for the excitation energies at Q=(0,0.75,0) are presented in Fig. 3 by the group of three vertical thin arrows above the scans. The excitations of the cone structure also cannot explain the inelastic scattering observed at low energies with increasing field.

In the case of quantum chains the field produces some spins aligned parallel to the field direction and approximately regularly distributed along the chain. If there is no correlation between the positions of the aligned spins in neighbouring chains, no long-range order will be formed even in the presence of a small interchain coupling. The IFP thus has incommensurate-like short-range order and the excitations form a spinon-magnon continuum according to calculations of Talstra and Haldane for the $1/r^2$ model, and Müller *et al.* for the nearest-neighbor model as shown schematically in Fig. 1(b). Measurements on Cs_2CuCl_4 for $B >$1.66 T are consistent with the absence of long-range antiferromagnetic or incommensurate ordering of the IFP phase. The energies for which the inelastic neutron scattering is expected to be large for 1D chains with an exchange interaction $\tilde{J}$, according to calculations

of Müller *et al.* [5], are shown in Fig. 3 by the thick vertical arrows above the scans. The observed splitting of the inelastic scattering and its behavior in a field is well reproduced by the calculation. The strong field dependence of the low-energy scattering results from the field dependence of the short-range ordering wavevector as it varies across the zone with increasing field. The experiments are consistent with the total scattering being constant for $B_C < B < 3$ T, in agreement with numerical simulations on finite quantum chains using the Müller Ansatz [5]. The regions where high intensity is expected in a constant-E scan are represented by horizontal bars in Fig. 4 and the measurements at the magnetic zone center Q=(-0.25,2,0) are also consistent with the calculated behavior.

In conclusion, we have measured the magnetic ground state and the excitations in the quasi-1D $S = 1/2$ HAF Cs_2CuCl_4 as a function of applied field. In the intermediate-field region there is no long-range antiferromagnetic or incommensurate ordering and the excitations are in agreement with predictions of Müller *et al.* for the 1D $S = 1/2$ HAF and Talstra and Haldane for the related $1/r^2$ chain. Mean field ordering and linear spin-wave theory cannot account for the observed behavior. Full details of these and related experiments on Cs_2CuCl_4 will be reported elsewhere [12].

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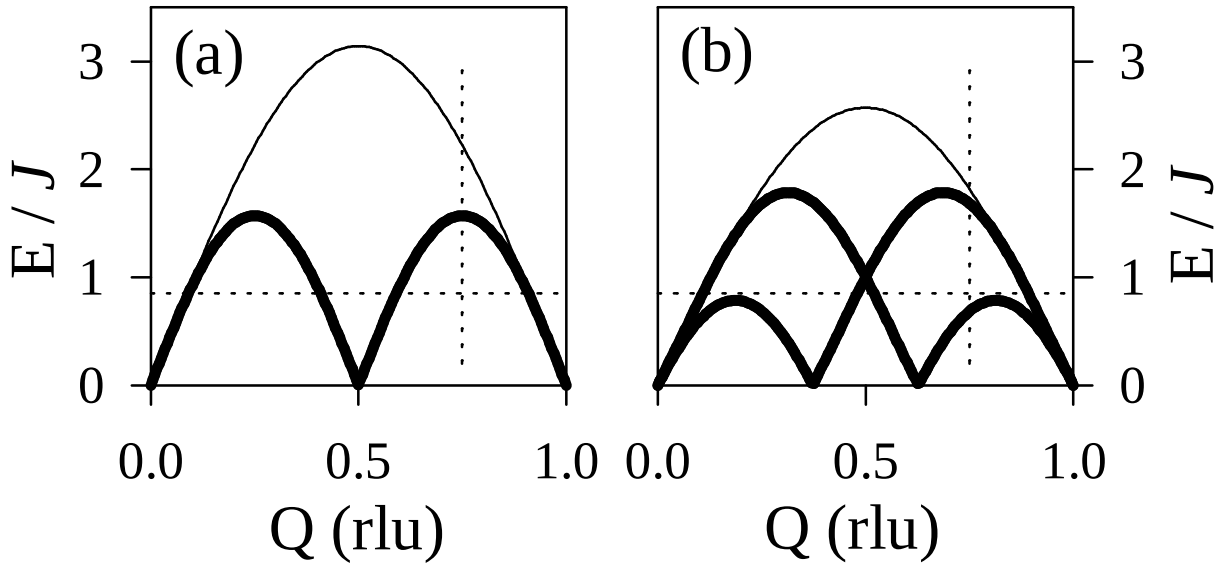
FIGURES

FIG. 1. Spectrum of longitudinal excitations in the 1D S=1/2 HAFC in (a) zero and (b) an applied field $B = J/g\mu_B$ [5]. The heavy and light lines are the boundaries of the scattering continua with the heavy lines indicating where strong scattering is expected. The dotted vertical and horizontal lines show the directions of constant-Q and constant-E scans in Figs. 3 and 4, respectively. The transverse modes also show similar effects, but have been omitted for the sake of clarity [5,6].

FIG. 2. (a) Magnetic chains and exchange paths in Cs_2CuCl_4. (b) Magnetic phase diagram in a field applied along c. The squares represent the measured boundary of the 3D cycloidal phase. The Intermediate Field and Ferromagnetic labels strictly refer to the $T = 0$ K line, but are effectively good descriptions of the ground state at very low temperatures $T \ll J/k_B$. (c) Dispersion of the magnetic excitations along the chain direction at $B = 0$ T. The solid line is a fit to the linear spin-wave dispersion.

FIG. 3. The intensity observed at Q=(0,0.75,0) as a function of energy and field. Two different scales are used and scans are shifted upwards by 100 (0 T)(right axis), 160 (1.66 T) and 80 (2 T)(left axis). Solid lines are fits as described in the text, and dotted lines represent the non-magnetic background. The energies at which the intensity is expected to be large for quantum chains [5] are shown by thick vertical arrows above the scans, while the thin arrows indicate the spin-wave energies for a cone structure. The horizontal bar in the zero-field scan represents the energy resolution (FWHM).

FIG. 4. The intensity observed for E=0.35 meV as a function of wavevector and field. Two different scales are used and scans are shifted upwards by 110 (0 T) (right axis), 150 (3 T) and 70 (4 T)(left axis). Solid lines are guides to the eye and dotted lines represent the non-magnetic background. Horizontal bars above the scans show where high intensity is predicted for quantum chains [5].


(a)
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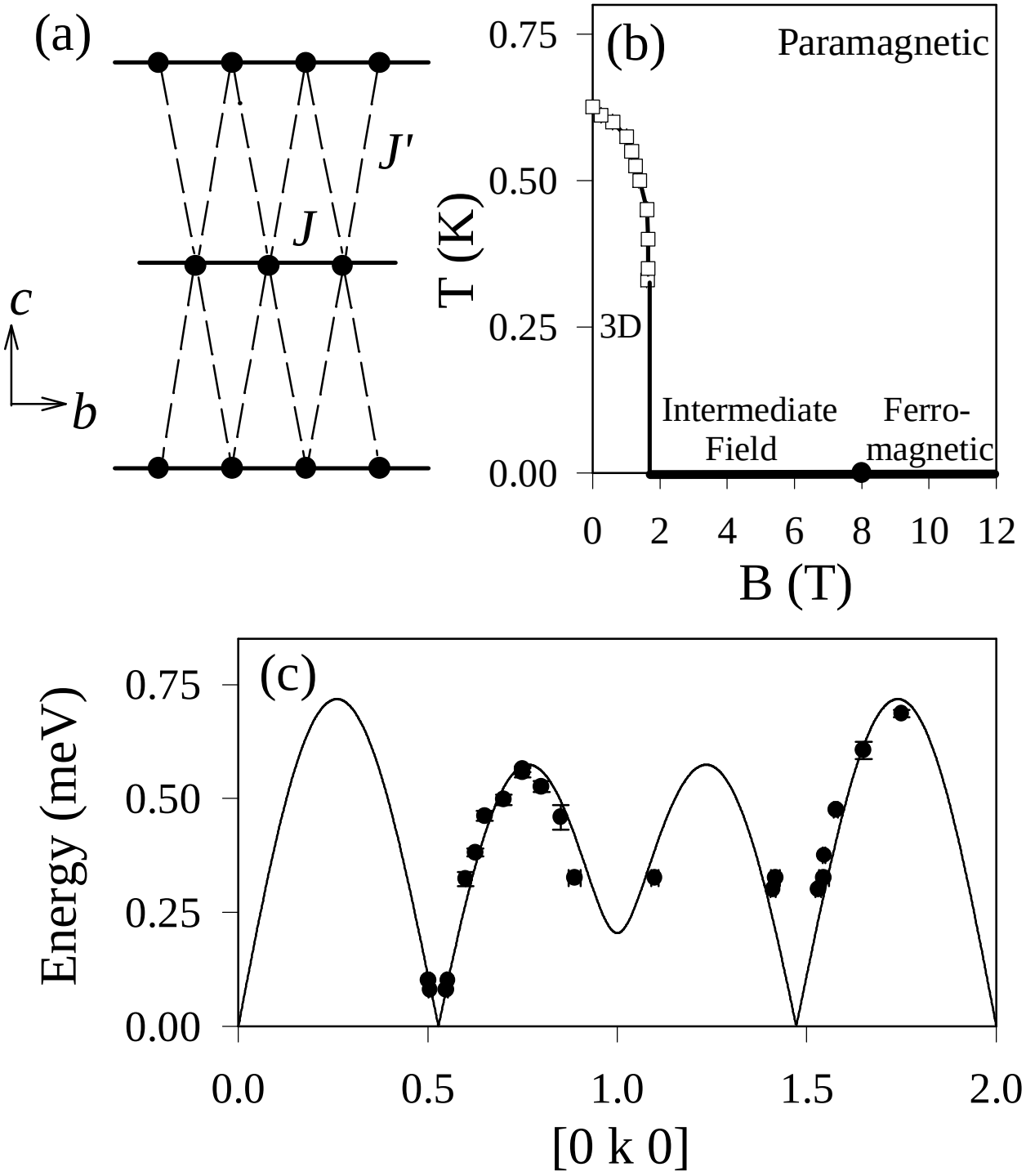
(a)
J'
J
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T (K)
3D
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0
2
4
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12
B (T)
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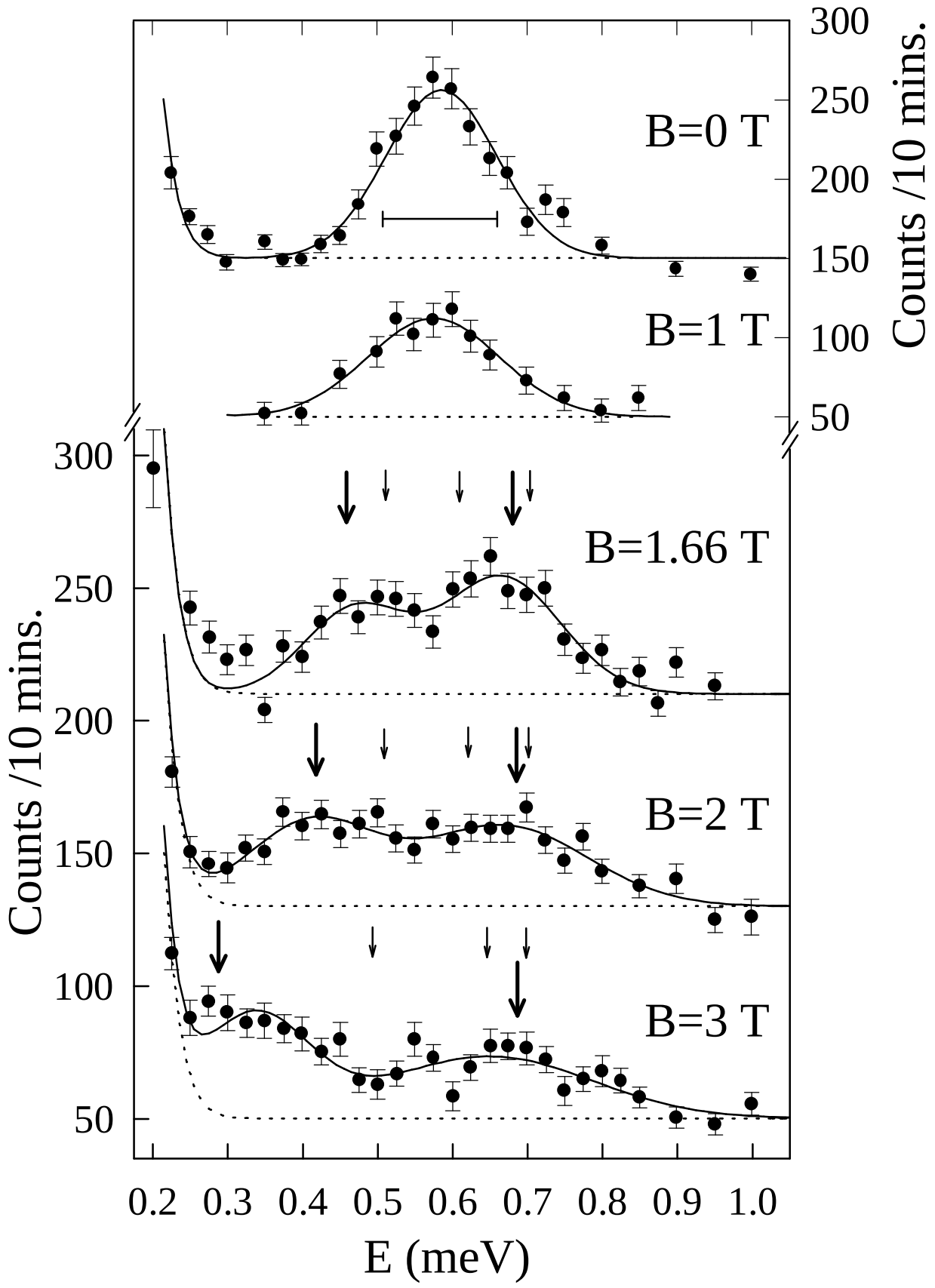
B=0 T
B=1 T
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Counts /10 mins.
E (meV)
300
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200
150
100
50
0.2
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0.8
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1.0

R. Coldea *et al.*, PRL, Figure 4